FAST UNCOOLED BiPbSrCaCuO DETECTORS

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ABSTRACT

Photoresponse of BiPbSrCaCuO superconductor films was studied at normal state temperatures. At room temperature, responsivity in the range of 10 - 100 V/MW was seen to remain constant over at least 8 orders of magnitude of incident power. Detectors were manufactured on films with a thickness as small as 20 nm, showing a time constant shorter than 10 ns. Investigation of photoresponse characteristics showed consistency with a tensorial Seebeck effect in off *c*-axis grown films. These characteristics suggest that uncooled BiPbSrCaCuO films may be useful replacements for pyroelectric power meters and photon drag detectors.

INTRODUCTION

Since the report of superconductivity in the multiphase BiSrCaCuO[1], much effort has been devoted to the film synthesis of this compound. One important motivation behind this effort is the potential of using high T_c superconducting films as radiation sensors. In our laboratories, BiSrCaCuO thin films were prepared using rf magnetron sputtering[2] and CO_2 laser ablation[3] techniques. Previously, the photoresponse of these films was studied at temperatures below the transition to the normal state. In addition to the bolometric detection mechanism,[4] a sub-gap nonbolometric mechanism was found to occur when the film was biased into a partially resistive regime.[5] In this work, the photoresponse was investigated specifically at normal state temperatures. This paper reports evidences of a fast nonbolometric mechanism in this temperature range. Characteristics of normal state photoresponse are presented, showing that uncooled BiSrCaCuO films may be useful replacements for pyroelectric power meters and photon drag detectors. Also, we discuss the consistency of these characteristics with a possible tensorial Seebeck effect in off *c*-axis grown films.

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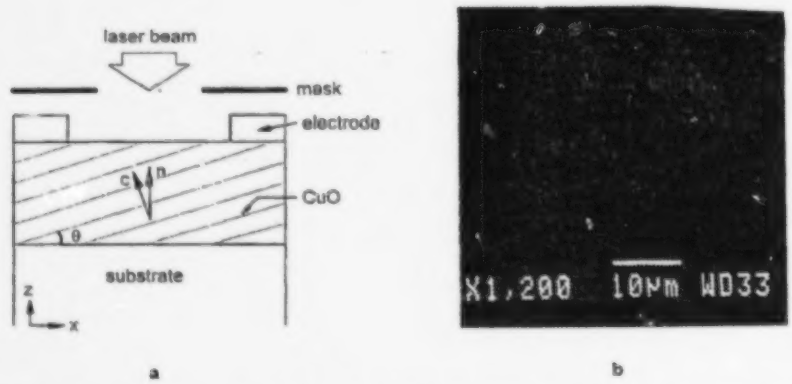

Figure 1. (*a*) Cross sectional schematic of a titled film with CuO planes inclined by an angle θ with respect to the substrate surface. A mask is used to prevent interaction between the incident beam and electrodes. (*b*) Scanning electron micrograph of a film detector used in the experiment.

EXPERIMENTAL

The BiSrCaCuO films studied in this work were prepared on (100) single crystal MgO, $LaAlO_3$, and $SrTiO_3$ substrates by rf magnetron sputtering. Pb was added to the films in cation ratio Pb:Bi:Sr:Ca:Cu = 1.2:2:2:2:3, as a precursor for the formation of the high T_c crystal structure. The film thicknesses ranged from 20 to 1000 nm. No intentional heating was applied to the substrates during the deposition. After a post-deposition annealing first in oxygen and then in air, films with a predominant $Bi_2Sr_2Ca_2Cu_3O_{10}$ crystal structure resulted. Details on the synthesis and structures of these films were reported elsewhere.[2] X-ray analyses showed that BiSrCaCuO films had a preferred *c*-axis orientation normal to the (100) plane of the substrate. To study effects of film orientation, MgO substrates specially cut with the surface at an angle $\theta \sim 10^0$ to the (100) plane were also used for film growth. The crystals grown on these substrates conserved the *c*-axis orientation normal to the MgO (100) plane, resulting in coherently tilted films with CuO planes inclined by 10^0 to the substrate surface (Fig. 1*a*). Typical films with an area of ~ 1 cm^2 was confirmed to be uniformly superconducting using a Faraday rotation magneto-optic technique.[6] Scanning electron microscopy revealed uniform morphological structure, small grain size, and no evidence of voids or domain boundaries (Fig. 1*b*). Some of the films were patterned into several strips using standard photolithography. Vacuum evaporation was used to form Ag electrodes on the films to be characterized. Films prepared with a thickness larger than 100 nm exhibited $T_c \sim 100$ K, which was consistent with the $Bi_2Sr_2Ca_2Cu_3O_{10}$ structure. However, T_c's ~ 80 - 90 K were obtained for films having a thickness smaller than 100 nm, indicating a transition towards the $Bi_2Sr_2CaCu_2O_8$ structure. This structural transition is believed to result from the low content of Pb in the thinner films.

The photoresponse was measured in the temperature range from T_c to 300 K. During the experiments, the film was thermally anchored to the cold finger of a standard liquid nitrogen cryostat. Temperature control was achieved by means of a Si thermometer, voltage regulating circuit, and heater in a feedback loop. The optical sources consisted of a 1.05-μm *Q*-switched Nd:YLF laser and a 10.6-μm TEA CO_2 pulsed laser. In some experiments the Nd:YLF source was operated in modulated cw mode. Photoresponses to pulsed and cw excitations were

measured with a 500-MHz oscilloscope and a lock-in amplifier, respectively.

RESULTS

Under laser illumination and without an externally applied bias current, a voltage signal $v(t)$ appeared across the two electrodes of the film. The magnitude of v was in the range of 10 - 100 V/MW, which was too large to be accounted for by conventional thermopower alone. This magnitude decreased by a factor of 2 when a resistor of resistance R was connected in parallel with the film resistance R, indicating a true current flow. We first shined a pulsed CO_2 laser beam between electrodes, 7 mm apart, using a moving opaque mask with a 2 x 1 mm^2 slit. The signal polarity remained unchanged as the laser beam scanned between electrodes, excluding the possibility that the signal was due to a differential temperature between ohmic contacts. Further support substantiating this observation is the fact that v decreased by a factor of ~ 10 when the laser beam was incident on each of the contacts. A polarity reversal was seen when the electrode positions were inverted, suggesting that v was induced by an electric field with a fixed orientation. To verify this, the spatial distribution of v in the film plane was measured. Several electrodes were placed in a circumferential area of 8.5-mm diameter on the film. The central area inside the circle was illuminated through a mask of 7-mm diameter. The voltages measured from diametrically facing electrodes showed a sinusoidal dependence on the angle formed by the line connecting the measuring electrodes and a fixed arbitrary axis. This voltage distribution suggests the creation of a field E with a component in the illuminated film area. The direction of E is fixed with respect to the film plane.

To determine the dependence of v on the active area of the film, mask slits of varying widths were successively used to scan a laser beam on a straight film line of equal segments A. Neglecting errors due to diffraction at the slit, a linear relation $v(\sum A) = \sum v(A)$ could be corroborated. The linear increase of v with the length of the line indicates also an uniform spatial distribution of the field in the illuminated area.

The temperature dependence of v was investigated. In this experiment, the Nd:YLF laser source was used in the low power cw mode (~ 240 mW) to avoid film heating. Figure 2*a* shows the temperature dependences of v and φ, where φ is the phase between voltage signal and laser modulation signal. When T was decreased from 300 to 160 K, v decreased continuously to a first minimum value. As T decreased further, v increased to attain a maximum near 110 K, and decreased to a second minimum near 90 K. No voltage signal was detected at temperatures below T_c. To each minimum of v an abrupt phase reversal was observed in the φ-T characteristic. There appears to be a relationship between the 160 K phase reversal and the 160 K anomaly observed in ultrasonic studies[7] of BiPbSrCaCuO superconductors. The origin of this anomaly is not well understood; a lattice structure transformation at specific temperatures might be responsible of this anomaly. The phase reversal, observed at at least 2 different temperatures, may be indicative of lattice instabilities if the origin of $v(t)$ is inherent to crystal structures. The R-T characteristic of the film is also shown in Fig. 2*a*. The temperature dependence of $\partial R / \partial T$ derived from this characteristic differs significantly from the temperature dependence of v, suggesting that v is nonbolometric in origin.

In the presence of a dc bias current i, a change of the laser induced voltage V_t was seen when i was inverted. This observation suggests a current independent component v contributes to the overall voltage, that is, $V_t = Ri + V + v$ where $V = i\,(\partial R / \partial T)\,\delta T$ is the bolometric component. Neglecting the dc component Ri and measuring the ac components of V_t with and without current inversion, v could be computed as $[V_t(i) + V_t(-i)] / 2$. We verified that the

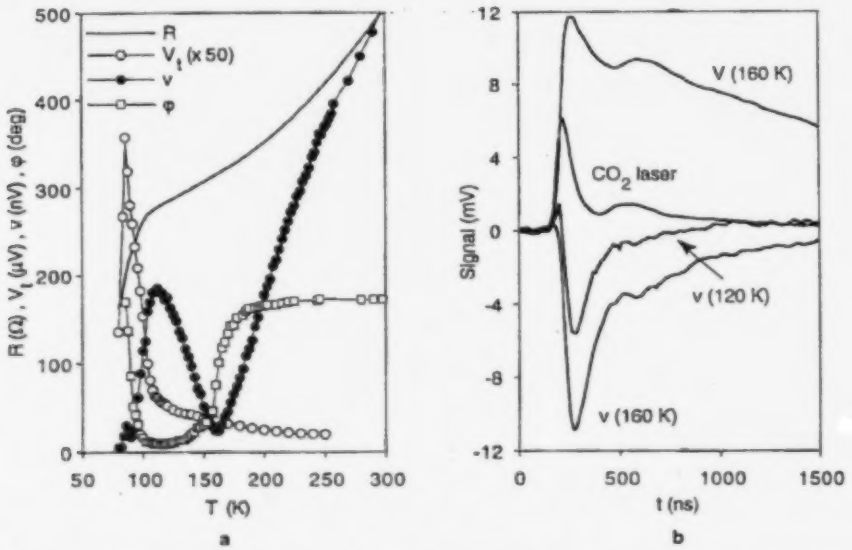

Figure 2. (*a*) Temperature dependences of R, V_t, v, and φ of BiPbSrCaCuO films; V_t was multiplied by a factor of 50 for visual aids. (*b*) Transient structures of incident CO_2 laser pulse, bolometric response V at $T = 160$ K, and current independent photoresponses v at $T = 120$ K and $T = 160$ K.

computed voltage signal reproduces the photoresponse measured in the absence of i. This result showed that the current independent response $v(t)$ and the bolometric response $V(t)$ superimpose linearly. The time averaged values of the sum signal V_t are plotted as a function of temperature in Fig 1*a*. Because the bolometric response is large near T_c, the temperature dependence of V_t is comparable with that of $\partial R / \partial T$ near the resistance transition. Figure 2*b* displays $v(t)$ and $V(t)$ together with the incident laser pulse. Here, $V(t)$ was singled out as $[V_t(t) - v(t)]$ and the incident pulse was recorded using a Ge photon drag detector. At $T = 160$ K, the half-amplitude fall times of $v(t)$ and $V(t)$ were measured to be 120 and 1220 ns respectively. The fact that the time constant of $v(t)$ is considerably shorter than the bolometric time constant supports the nonbolometric origin of v. Another remarkable feature is the phase reversal of the voltage signal due to laser heating. Because of the high intensity of the CO_2 laser pulse used in this experiment (~ 5 mJ), the temperature change of the film during illumination may be large (~ 100 K). In Fig. 2*b*, a phase reversal in the transient response $v(t)$ of a film biased at $T = 120$ K is seen. Referring to Fig. 2*a*, the occurrence of this phase reversal could be anticipated as the transient temperature of the film reached 160 K after a short period of laser heating. The transient phase reversal may provide useful information on the transient temperature of the film for different periods of laser heating.

DISCUSSION

Results of a series of experiments evidenced the thermal origin of the nonbolometric normal state response. It was observed that similar response characteristics could be obtained when the laser source was replaced with a heat source. Secondly, the normal state response is

inherently independent of radiation wavelength; measurements of v in the spectral range from 0.6 to 12 μm showed a dependence comparable with the spectral absorptance[4] of the film. Among possible nonbolometric thermal mechanisms, pyroelectricity, piezoelectricity, and thermoelectricity were considered. In experiments where a cw source provided continuous heating to the film, a dc voltage could be maintained across the film, excluding the possibility of pyroelectricity. Piezoelectricity was excluded as a possible mechanism from the observation that the response was independent of the light polarization. Thermoelectricity results in a field $E = S\,\nabla T$ due to the Seebeck effect, where S is the Seebeck tensor and ∇T the temperature gradient. Assuming unidimensional heat flow in the direction normal to the substrate surface, the transverse component of this field can be expressed as $E_x = S_{xz}\,(\partial T / \partial z)$, where S_{xz} is the offdiagonal tensorial element of S. It is believed that, due to misalignment in cutting of substrates or film growth conditions, off-c axis grown films were generally obtained. In this case, referring to the coordinates specified in Fig. 1*a*, the offdiagonal element $S_{xz} = (S_{ab} - S_c) \sin\theta\cos\theta$ has a finite value, giving rise to a transverse voltage $v = E_x x_0 \sim (S_{ab} - S_c)\,\Delta T\,(x_0/\Delta z) \sin\theta\cos\theta$. Considering the difference $(S_{ab} - S_c) \sim -1.2$ μV/K of absolute thermopowers in the ab plane and along the c axis of BiSrCaCuO single crystal,[8] a normalized voltage $v / \Delta T \sim 82$ mV/K is obtained for a film with a thickness $\Delta z = 20$ nm, titled by $\theta = 10^0$ and illuminated across $x_0 = 8$ cm. This estimation shows that large transverse thermoelectric voltages may be generated when radiation induces a small temperature difference between film surface and film bottom. In support to the offdiagonal thermoelectric origin of the signal, responsivity was seen to increase by a factor of ~ 2 for detectors prepared on MgO substrates specially cut with $\theta = 10^0$. Given $v(\theta_1) / v(\theta_2) \sim \theta_1 / \theta_2$ for small values of θ, a factor of 2 implies that films grown on standard (100) substrates be titled by ~ 5^0. This misalignment can reasonably be speculated considering the size of possible imperfections of the substrate surface.

Because the thermoelectric response is proportional to the temperature difference between film surface and film bottom, its time constant is determined by the diffusion time $\tau \sim (\Delta z)^2 / D$ for heat propagation through the film. Using a thermal diffusivity constant $D \sim 2 \times 10^{-3}$ cm²/s for room temperature BiPbSrCaCuO,[9] τ could be estimated for different values of film thickness. In Table 1, the computed time constant is compared with the fall time (from 90% to 10% of signal amplitude) of the transient response of the film to a Nd:YLF laser pulse. Good agreement between the computed and measured time constants was obtained over an extended range of film thickness, corroborating the suggestion that the response is thermoelectric in origin. The fall time measured on the 20-nm thick film may be limited by the fall time of incident excitation; using a picosecond InGaAs detector, the fall time of the Nd:YLF laser pulse was measured to be ~ 7 ns.

Table 1. Computed and measured time constants of the thermoelectric response for different thicknesses of BiPbSrCaCuO detectors.

	Thickness (nm)	Computed τ (ns)	Fall time (ns)
Detector	20	2	8
	50	13	12
	100	50	55
	150	113	115
	300	450	430
	430	925	890
Laser pulse			7

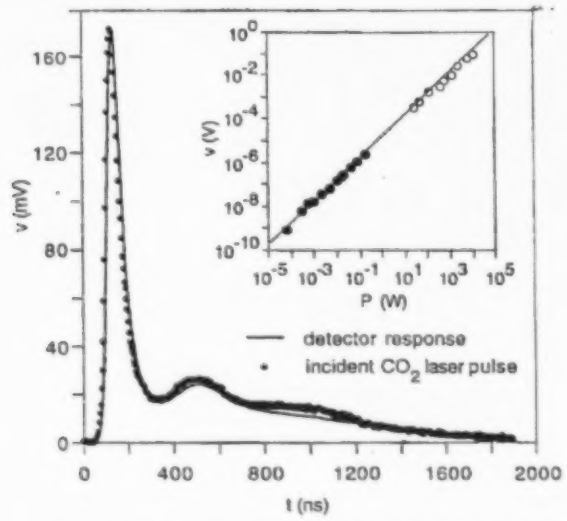

Figure 3. The transient response of a 20-nm thick detector reproduces the transient structure of an incident TEA CO_2 laser pulse, showing a short time constant. The inset shows the dynamic range of at least 8 orders of magnitude obtained on the same detector. The solid line depicts a slope of unity. Open circle and solid dot data were obtained using an Nd:YLF laser source respectively in pulsed and cw modes.

The short time constants measured at the near infrared wavelength could further be obtained at the long wavelengths, suggesting that the effect of optical penetration depth may be small. Figure 3 displays the response of a detector manufactured on a 20-nm thick film to a short CO_2 laser pulse. Again, this response reproduces the transient structure of incident pulse, showing a short thermal time constant. The potential of achieving sub-nanosecond response time in thin BiPbSrCaCuO films makes them an attractive choice for pulsed laser characterization. The primary advantage of these detectors is their responsivity is more than 100 times that of photon drag detectors (~ 0.2 V/MW), which are traditionally used for this application. Secondly, unlike the drag detector, superconductor detectors exhibit responsivity which is intrinsically independent of the wavelength and direction of incident beam. This large, spectrally independent responsivity also makes them a useful replacement for pyroelectric power meters. Because of the nonuniform spectral response of pyroelectric power meters at far infrared wavelengths, it is difficult to calibrate and operate them in this spectral range. In previous studies, the spectral response of BiPbSrCaCuO detectors was shown to be relatively uniform for wavelengths above 8 μm.[4] In addition, a large dynamic range was measured for these detectors. The inset of Fig. 3 shows a linear increase of voltage response over at least 8 orders of magnitude of incident power for a detector with a film thickness of 20 nm. In this experiment, the range of incident power was limited by the range of our Nd:YLF laser source which operated in the low power cw mode and the high power pulsed mode. The continuity of the data sets obtained by cw radiation heating and pulsed radiation heating confirmed that the time constant of the detector is equal to or shorter than the time constant of the incident excitation.

CONCLUSIONS

The photoresponse characteristics of normal state BiPbSrCaCuO films were reported. At room temperature, responsivity in the range of 10 - 100 V/MW was obtained. This large responsivity was shown to be consistent with a tensorial Seebeck effect in off *c*-axis grown films. In support to this effect, good agreement was found between the time constant of film detectors and the diffusion time for heat propagation through the film. Detectors were manufactured on films with a thickness as small as 20 nm, showing a time constant shorter than 10 ns. A dynamic range of at least 80 dB was measured for these detectors. The large responsivity, speed, dynamic range, and spectral range of uncooled BiPbSrCaCuO detectors suggest that they may be useful replacements for pyroelectric power meters and photon drag detectors. Furthermore, because the thermoelectric response is inherent to the crystal structure of the film, it can be used to probe changes of film structure at specific temperatures.

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REFERENCES

1. H. Maeda, Y. Tanaka, M. Fukutomi, and T. Asano, A new high-T_c oxide superconductor without a rare earth element, *Jpn. J. Appl. Phys.* 27:L209 (1988).
2. L. Ngo Phong, B. Tremblay, I. Shih, and C. X. Qiu, Accelerated formation of the high Tc phase in moderately Pb doped BiSrCaCuO thin films, *Supercond. Sci. Technol.* 5:555 (1992).
3. A. L. Li, I. Shih, and A. A. Gundjian, CO_2 laser ablated BiSrCaCuO superconducting thin films, *Physica C* 208:64 (1993).
4. L. Ngo Phong, I. Shih, and P. Laou, IR reflectance and responsivity of PbBiSrCaCuO films, *SPIE* 2269:131 (1994).
5. L. Ngo Phong and I. Shih, Photoresponses of granular (Pb)BiSrCaCuO films to millimeter wavelength radiation, *J. Appl. Phys.* 74:7414 (1993).
6. E. Batalla, E. G. Zwartz, R. Goudreault, and L. S. Wright, Color imaging the magnetic field distribution in superconductors, *Rev. Sci. Instrum.* 61:2194 (1990).
7. P. Venugopal Reddy and S. Shekar, Ultrasonic studies of some Bi-based high-T_c superconductors, *Supercond. Sci. Technol.* 6:785 (1993).
8. M. Pekala, K. Kitazawa. A. M. Balbashov, A. Polaczek, I. Tanaka, and H. Kojima, Anisotropic thermoelectric power and thermal conductivity in superconducting single crystals BiSrCaCuO, *Solid State Commun.* 76:419 (1990).
9. J. Isaac, J. Philip, M. T. Sebastian, and A. D. Damodaran, Thermal Diffusivity of BiSrCaCuO superconductor - effect of Pb doping, *Physica C* 199:247 (1992).